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ORIGINAL PAPER

Copper and zinc concentrations of medicinal herbs and soil surrounding ponds on agricultural land

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Abstract This study measured copper and zinc concentrations in medicinal herbs and soil sampled from three agricultural fields with ponds from mid-June till the end of July 2014. Six herb species were tested: *Potentilla anserina* L., *Mentha arvensis*, *Achillea millefolium* L., *Comarum palustre* L., *Lysimachia vulgaris* L., and *Lycopus europaeus* L. Two of the ponds were in the borough of Jabłonna Lacka and one in the borough of Sabnie. The fields around each pond were divided into transects with three concentric soil moisture sections, each with different soil water contents. Soil and plant samples were taken from each transect to determine the concentrations of copper and zinc by inductively coupled plasma atomic emission spectrometry. Despite the use of fertilizers and pesticides in the fields, copper and zinc had not polluted the soil or the plants growing in the soil depressions. Out of the six plants tested, only *C. palustre* had an increased concentration of zinc. Different farming methods used on the fields affected the concentrations of copper and zinc in the soil of the different depressions. The highest concentrations of these metals were found in the soil of the periodically wet sections.

Keywords Fertilizer · Pesticide · Heavy metal · Habitat loss · Contamination

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Introduction

The development of modern farming in Poland has contributed to the disappearance of many natural habitats, and areas of natural and semi-natural vegetation are currently decreasing. The contribution of small bodies of water in agricultural fields to environmental sustainability is currently of great interest (Nicolet et al. 2007; Boix et al. 2012). These bodies of water usually do not exceed 1 ha in area, but because of seasonal water table oscillations their water levels rise and fall.

In Poland, ponds in agricultural fields are usually located in postglacial areas within moraine plateaus and within moraine deposits. Plants growing in or near such bodies of water often have an excess of nutrients and heavy metals, both derived from agricultural fields. Fertiliser and pesticide use, together with tillage, result in the accumulation of heavy metals in depressions of agricultural fields. Yet the contents of macro- and microelements in plants growing there vary as a result of soil properties, environmental pollution, and climatic conditions (Chan 2003; Ražic et al. 2008; Zheljazkova et al. 2008).

Apart from their nutritional value, many herbs have antioxidant, antibacterial and anti-inflammatory properties, regulate digestion, and may also be used to preserve food (Krejpcio et al. 2007; Bielicka-Giełdoń et al. 2011). Unfortunately, despite the many health benefits of herbs, they can absorb pollutants harmful to living organisms. Medicinal and culinary herbs in Poland may be sourced from natural habitats, so their contents of heavy metals and other pollutants should be monitored to prevent their build-up in the organisms which consume them (Sembratowicz et al. 2009; Ulewicz-Magulska et al. 2009; Luginina and Egoshina 2013). The use of medicinal herbs is currently increasing, thus the monitoring of herbal materials should be more systematic, especially for medicinal plants sourced from the wild (Deng et al. 2004; Garcia-Rico et al. 2007; Arslan et al. 2010). For example, despite their important functions in human metabolism, copper and zinc can be toxic when ingested at high doses (Gaetke and Chow 2003; Tapiero and Tew 2003; Yruela 2005; Sahoo et al. 2010).

The aim of this experiment was to determine the contents of copper and zinc in herbs sampled from three transects of soil with different moisture contents, around three in-field ponds.

Materials and methods

Sampling was undertaken in experimental areas of the Siedlce Plateau from mid-June till the end of July 2014. Six species of herbs were selected which grew around in-field ponds: silverweed (*Potentilla anserina* L.), corn mint (*Mentha arvensis*), yarrow (*Achillea millefolium* L.), purple marshlocks (*Comarum palustre* L.), yellow loosestrife (*Lysimachia vulgaris* L.) and gypsy-wort (*Lycopus europaeus* L.). Three ponds without outlets were used in the experiment: two in the village of Bujały Mikoszethe in the borough of Jabłonna Lacka (pond 1 and pond 2), and one in Grodzisk in the borough of Sabnie (pond 3). All three ponds were situated in Sokołów Podlaski County, in the eastern part of Mazovian Voivodeship.

The ponds were selected according to the following criteria: location in agricultural land, diversity of vegetation, size ranging from 15,000 (pond 3) to 850 m^2 (pond 1). Pond 1 was surrounded by an agricultural field planted to cereals, pond 2 was surrounded by permanent grassland, and pond 3 was overgrown with bushes in a field sown to cereals which were at a distance of 80 m. The ponds were typical for the area, situated in clearly visible depressions, permanently or periodically filled with water and overgrown with vegetation typical for wetlands (Skwierawski 2005). The soils were defined as luvisol and rusty types. The experimental area around each pond was divided into three 18- to 80-m-long transects, from the edge of the water to the edge of the field. The transects were divided into three sections according to their moisture contents: a wet section with water stagnating in early spring after the snow has melted or after heavy rainfall; a periodically wet section with water stagnating only in the early spring after the snow has melted; a dry section with no water stagnation (Franczak and Franczak 2015). Plant and soil samples, 85–100 samples from each area, were taken from the concentric sections. Bottom sediment samples were taken from each pond.

The plant material was ground to give 0.25-mm-diameter particles, weighed, and 1 g put into a porcelain crucible. The samples were incinerated in a muffle furnace at 450 °C for 15 h. Ten millilitres of hydrochloric acid solution (1:1) was then added and the plant material was put into a sand bath to decompose carbonates and to isolate silicon. The contents of the crucible were filtered through a hard filter into a 100-ml conical flask. Copper and zinc concentrations were determined by inductively coupled plasma-atomic emission spectrometry. The pH of the soil and the sediment was measured by the potentiometric method; the pH varied from 5.45 to 6.50. The experimental factors were plant species, the moisture sections they grew in, and the type of field.

The means were analysed by ANOVA (Statistica, version 10.0; StatSoft). Tukey's test was used to calculate the least significant difference at $p < 0.05$. For each moisture zone, the correlation coefficient between the total contents of copper and zinc in the plant biomass and in the soil were also determined.

Results and discussion

Copper concentrations in the biomass of the test plants ranged from 4.69 to 12.47 mg kg^{-1} dry mass (DM) and varied significantly, depending on the species and the moisture section where they grew (Table 1). When taking into account all the moisture sections, the highest concentration of copper was found in *M. arvensis* (10.07 mg kg^{-1}) and *A. millefolium* (9.99 mg kg^{-1}), and the lowest in *L. vulgaris* (5.26 mg kg^{-1}). No significant differences were found between the sections in the average plant concentrations of copper, which varied from 8.14 mg kg^{-1} in the periodically wet sections to 8.28 mg kg^{-1} in the dry sections. However, there were significant differences in the plants growing next to each pond. The highest copper concentrations were found in herbs growing near the pond surrounded by permanent grassland, in the periodically wet sections (8.86 mg kg^{-1}). The concentrations of copper were lower in the other areas. For example, in the pond area surrounded by cereal crops and that surrounded by bushes, the concentrations of copper in plants growing in the wet sections were 8.42 and 8.13 mg kg^{-1}, respectively. Soil moisture affects enzyme activity and the redox potential of soil, which might have increased the concentrations of soluble forms of copper. According to Kabata-Pendias and Pendias (1999) a typical concentration of copper in plants ranges from 2 to 20 mg kg^{-1} DM, while a concentration >30 mg Cu kg^{-1} DM is toxic. In our experiment the concentration of copper did not exceed 30 mg Cu kg^{-1} DM, and only in some plants (*M. arvensis* and *A. millefolium*) was it slightly higher than 10 mg kg^{-1}. In a previous study, the concentration of copper in the leaves of *Taraxacum* sp. was 10.2 mg kg^{-1} (Królak 2001). Blicharska et al. (2008)

Table 1 Copper and zinc concentrations (mg kg^{-1}) in the biomass of six herb species

Area	Pond 1[a]				Pond 2[b]				Pond 3[c]				Mean			
Species	I	II	III	Mean	I	II	III	Mean	I	II	III	Mean	I	II	III	Mean
Copper																
Potentilla anserina L.	7.89bC	7.47bB	8.76aC	8.04C	6.16Eb	7.56Ca	8.63Ba	7.45D	7.98Cab	8.09Ba	7.56Db	7.88C	7.34Ba	7.71Ca	8.32Ba	7.79B
Mentha arvensis	10.57aA	9.56bA	10.23aB	10.12A	10.60Bb	11.98Aa	10.78Ab	11.12B	9.01Bab	9.34Aa	8.54Cb	8.96B	10.06Aa	10.29Aa	9.85Aa	10.07A
Achillea millefolium L.	9.62aB	7.56cB	8.57bC	8.58B	11.98Aab	12.47Aa	11.38Ab	11.95A	9.87Aa	8.18Bb	10.24Aa	9.43A	10.49Aa	9.40Bb	10.06Aab	9.99A
Comarum palustre L.	6.08aC	5.36bD	6.12aD	5.85D	7.02Dab	6.32Db	7.59Ca	6.98E	6.02Db	7.41Ca	6.99Ea	6.81D	6.37Ca	6.36Da	6.90Ca	6.55C
Lysimachia vulgaris L.	5.89bC	6.48aC	4.69cE	5.69E	4.98Fa	5.11Ea	4.78Da	4.96F	5.69Ea	5.02Db	4.69Fb	5.13E	5.52Da	5.54Ea	4.72Da	5.26D
Lycopus europaeus L.	10.47aA	9.87bA	10.85aA	10.40A	8.45Cb	9.72Ba	9.03Bab	9.07C	10.23Aa	8.99Ac	9.63Bb	9.62A	9.72Aa	9.53ABa	9.84Aa	9.70A
Mean	8.42a	7.72c	8.20b	–	8.19b	8.86a	8.70a	–	8.13a	7.84b	7.94b	–	8.25a	8.14a	8.28a	–
Zinc																
P. anserina	40.78Db	45.39Cb	58.04Ca	48.07D	29.95Ec	42.31Ca	40.21Db	37.49D	50.12Ca	33.79Dc	40.23Db	41.38D	40.28Db	40.50Db	46.16Da	42.31C
M. arvensis	63.10Ca	47.23Cb	50.23Db	53.52C	50.78Ca	37.01Dc	45.21Cb	44.33C	80.47Bb	85.63Ca	70.23Cc	78.78B	64.78Ca	56.62Cb	55.22Cc	58.88B
A. millefolium	27.32Fa	28.36Da	20.11Eb	25.26E	23.01Fb	28.12Ea	27.85Ea	26.33F	30.11Da	25.16Eb	23.78Fc	26.35E	26.81Fa	27.21Ea	23.91Fb	25.98D
C. palustre	101.3Ac	114.8Aa	109.4Ab	108.5A	99.87Ac	103.8Ab	118.7Aa	107.5A	120.3Ab	109.9Ac	122.6Aa	117.6A	107.2Ac	109.5Ab	116.9Aa	111.2A
Lysimachia vulgaris	30.28Ea	21.89Eb	24.41Eb	25.53E	36.32Da	25.98Fb	21.49Fc	27.93E	20.14Ec	24.89Eb	29.98Ea	25.00F	28.91Ea	24.25Fc	25.29Eb	26.15D
Lycopus europaeus	85.12Ba	83.69Bb	76.12Bb	81.64B	70.11Bb	74.28Ba	75.55Ba	73.31B	81.02Ba	79.00Bb	73.09Bc	77.70C	78.75Ba	78.99Ba	74.92Bb	77.55E
Mean	57.98a	56.89a	56.39a	–	51.67b	51.92b	54.84a	–	63.69a	59.73b	59.99b	–	57.78a	56.18c	57.07c	–

Within a row, *different lowercase letters* indicate a significant difference; *within a column*, *different uppercase letters* indicate a significant difference

I Wet section, *II* periodically wet section, *III* dry section

[a] Cultivated field

[b] Permanent grassland

[c] Area with bushes

found the highest concentration of copper in herbs available on the Polish market in hawthorn berries (16.4 mg kg^{-1}), and the lowest in eyebright (1.77 mg kg^{-1}). With the exception of the pond within a cultivated field, the concentration of zinc in the plants varied significantly, according to both species and moisture section (Table 1), and ranged from 20.11 to 122.6 mg kg^{-1} DM. Two species, *C. palustre* and *L. europaeus*, had the highest average concentrations of zinc (111.2 and 77.55 mg kg^{-1}, respectively), and the lowest average concentrations were found in *A. millefolium* (25.98 mg kg^{-1}) and *L. vulgaris* (26.15 mg kg^{-1}). In the wet sections, the highest average concentrations of zinc were in *M. arvensis* (64.78 mg kg^{-1}) and *L. vulgaris* (28.91 mg kg^{-1}). In the periodically wet sections the highest concentrations of zinc were in *A. millefolium* (27.21 mg kg^{-1}) and *L. europaeus* (78.99 mg kg^{-1}).

In the dry sections, *P. anserina* (46.16 mg kg^{-1}) and *C. palustre* (116.9 mg kg^{-1}) had the highest concentrations of zinc. The average concentration of zinc for all the herbs species was 57.02 mg kg^{-1}. However, according to Ulewicz-Magulska et al. (2009) herb material contains 44.82 mg Zn kg^{-1} on average. The highest accumulation of zinc was found in plants growing in the wet sections, then in the dry sections, and the lowest in the periodically wet sections. The concentration of this metal varied also according to the experimental area. The highest concentration of zinc (61.14 mg kg^{-1}) was found in herbs growing by the pond surrounded by bushes and the lowest (52.82 mg kg^{-1}) by the pond surrounded by a cultivated field.

The natural concentrations of zinc in plants growing in Poland vary depending on the species, and ranges from 10 to 50 mg kg^{-1} DM (Kabata-Pendias 2002). In an experiment carried out in the south of the Podlasie Voivodeship, the average concentration of zinc in the leaves of *Taraxacum* reached 65.8 mg kg^{-1} (Królak 2001). In Lower Silesia Voivodeship the concentration of zinc was 32.63 mg kg^{-1} on average in medicinal plants grown in fields and destined for the herb industry (Wiechuła et al. 2013). Suchacz and Wesołowski (2012) found that, in different groups of medicinal plants, the zinc concentration ranged from 8.62 to 263.67 mg kg^{-1}. A concentration of zinc >100 mg kg^{-1} DM may cause developmental disorders of plants (Kabata-Pendias and Pendias 1999).

The concentrations of copper and zinc in the bottom sediment of the three ponds varied (Table 2). The highest concentrations of copper (7.71 mg kg^{-1}) and zinc (77.1 mg kg^{-1}) were found in the bottom sediment of the pond within a cultivated field. In the sediment from the pond surrounded by bushes, the concentration of copper was five times lower, at 1.42 mg kg^{-1}, while the concentration of zinc was eight times lower, at 9.06 mg kg^{-1}. This might have been a result of the use of fertilisers and pesticides, as well as tillage, which contributes to water and wind erosion.

For all soil samples, the highest concentrations of both metals were found in those from the periodically wet sections (Table 3). The concentration of copper in the soil here was 3.20 mg kg^{-1} on average, while in the wet sections it was 2.74, and in the dry sections 3.04 mg kg^{-1}. The average concentration of zinc in the periodically wet sections was 28.31 mg kg^{-1}, and it was significantly lower in the other sections, i.e., 23.10 mg kg^{-1} in the wet sections, and 13.41 mg kg^{-1} in the dry sections. In the soil around the pond surrounded by permanent grassland there was nearly twice as much zinc and copper as in the soil around the pond with bushes.

Plants growing in the field with permanent grassland accumlated the most copper (Table 1). The content of heavy metals in soil is not always related to that of the plants which grow on it (Aydinalp and Marinova 2003), and despite the fact that the lowest concentration of zinc was found in the bottom sediment and in the soil in the area

Table 2 Concentrations of copper and zinc in the bottom sediments of the ponds [mg kg^{-1} dry matter (DM)]

Metal	Pond 1	Pond 2	Pond 3	mean
Cu	7.71	6.60	1.42	5.20
Zn	77.1	45.60	9.06	43.90

For ponds, see Table 1

Table 3 Concentrations of copper and zinc in different moisture sections of the soil (mg kg^{-1} DM)

Area	Copper				Zinc			
	I	II	III	Mean	I	II	III	Mean
Pond 1	1.81Ba	4.30Aa	2.00Ba	2.70A	20.40Bb	34.51Aa	13.50Bc	22.80B
Pond 2	4.50Aa	3.40Ab	3.71Ab	3.87C	38.91Aa	35.20Ab	15.81Ac	29.97A
Pond 3	1.91Ba	1.90Ba	2.70Bb	2.17B	10.00Cb	15.22Ba	10.93Cb	12.05C
Mean	2.74a	3.20a	3.04b	–	23.10b	28.31a	13.41c	–

Within a row, different lowercase letters indicate a significant difference; within a column, different uppercase letters indicate a significant difference

For abbreviations, see Tables 1 and 2

Table 4 Linear correlation coefficients between plant biomass and soil copper concentrations

Species	Soil	*P. anserina*	*M. arvensis*	*A. millefolium*	*C. palustre*	*Lysimachia vulgaris*	*Lycopus europaeus*
P. anserina	−0.281	1.00					
M. arvensis	0.399	−0.117	1.00				
A. millefolium	0.196	−0.417	0.618*	1.00			
C. palustre	−0.122	−0.033	0.038	0.474	1.00		
Lysimachia vulgaris	0.139	−0.119	−0.127	−0.487	−0.777*	1.00	
Lycopus europaeus	−0.176	0.554	−0.146	−0.519	−0.753*	0.375	1.00

* $p \leq 0.05$; $n = 12$

Table 5 Linear correlation coefficients between plant biomass and soil zinc concentrations

Species	Soil	*P. anserina*	*M. arvensis*	*A. millefolium*	*C. palustre*	*Lysimachia vulgaris*	*Lycopus europaeus*
P. anserina	−0.052	1.00					
M. arvensis	0.522	−0.084	1.00				
A. millefolium	−0.019	−0.074	0.059	1.00			
C. palustre	−0.121	−0.054	0.095	0.161	1.00		
Lysimachia vulgaris	0.215	−0.592*	−0.123	−0.485	0.276	1.00	
Lycopus europaeus	0.040	0.394	−0.287	0.453	0.523	−0.474	1.00

* $p \leq 0.05$; $n = 12$

with bushes, the accumulation of this metal by plants was highest here (Table 1). The differences in the heavy metal concentrations in the soils of the depressions result from different tillage methods used in the fields surrounding them and from the phytoremediation properties of the plants growing there (Cataldo and Wildung 1978; Ernst 1996; Raskin et al. 1997). Metal solubility and bioavailability are usually related to the soil contents of organic matter, clay minerals, oxides of metals such as iron, aluminium and manganese, and soil pH (Kabata-Pendias 2002; Burzyńska 2009; Domańska 2009). Plant species is a deciding factor in the accumulation of zinc and other heavy metals (Lock and Janssen 2003; Rosselli et al. 2003; Kwiatkowska-Malina and Maciejewska 2011). The contents of zinc and copper in this experiment did not exceed the limits for soil contamination in Poland (Kabata-Pendias and Pendias 1999).

The linear correlations between the concentrations of copper and zinc in the soil and their concentrations in the plants were not statistically significant (Tables 4, 5). However, there was a significant relationship between the concentrations of copper in *A. millefolium* and *M. arvensis,* and a negative relationship between those of *Lysimachia vulgaris* and *Lycopus europaeus* on the one hand and *C. palustre* on the other. There was a negative correlation between the zinc concentrations in *L. vulgaris* and *P. anserina*. The concentrations of copper and zinc in the herbs varied significantly according to species and where the plants grew. Taking into account the average values of all areas and moisture sections, the highest concentrations of copper were in *M. arvensis* and *A. millefolium*; for zinc the highest concentrations were in *C. palustre* and *L. europaeus*. Out of all the analysed herbs, only in the samples of *C. palustre* was the concentration of zinc high, i.e. 100 mg kg^{-1}, a concentration which may be toxic for some plants. The concentrations of zinc and copper varied between the sediments from all the three experimental areas of the Siedlce Plateau, with the highest concentrations in the pond surrounded by arable land. The highest concentrations of copper and zinc, averaged for all three areas, were in the soil taken from the periodically wet sections. Soil from the field with permanent grassland had the highest concentration of these metals.

In conclusion, the use of pesticides and mineral fertilisers in the sampled areas did not cause the contamination of the plants growing there with zinc and copper. The plant and soil concentrations of these metals indicate that herbs can be grown and gathered from these areas for medicinal use.

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